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(54) Insecticidal compositions, and preparation and use thereof.

(57) An improved insecticidal particulate composition is a free-flowing composition comprising 1-50% by weight of methomyl impregnated in 10-98% by weight of a water-insoluble polymer which is soluble in an organic solvent, has an inherent viscosity of at least 0.2 and is capable of releasing methomyl at a rate such that said composition exhibits an undiminished initial activity, an enhanced residual activity and reduced phytotoxic damage to cotton plants relative to solid methomyl.

The composition may be made by precipitating the components from solution by comingling with a non-solvent under conditions of high shear.

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"Insecticidal compositions, and preparation
and use thereof"

This invention relates to compositions of the insecticide methomyl and to the preparation and use of such
5 compositions.

It has been recognized that particles of insecticide may be used effectively to control insects while simultaneously controlling the speed of release of the insecticide so as to minimize any undesirable
10 effects. Traditionally, the particles are produced by processes which involved co-melting the pesticides with a barrier which may be a polymer. After the polymer and insecticide have been melted together, a grinding step takes place in which the particles are reduced to the
15 desired size. This sort of technique is taught in Canadian Patent 51,153 to G. C. Allen, Coppedge et al., J. Economic Entymology 68, 508 (1976), and Canadian Patent 786,777.

Methomyl is an important insecticide with a
20 wide variety of uses. It is known chemically as S-methyl N-(methylcarbamoyloxythioacetamide). Although it has widely been used on a variety of crops to kill insects, without causing any significant damage to the crops, the use of methomyl has been restricted with
25 respect to cotton. In the past, although serving to control effectively the insects which affect cotton such as bollworms, budworms, cotton leaf perforater, beet armyworm and looper, the use of methomyl has re-

sulted in phytotoxicity. This is evidenced by reddening of the cotton. Although methomyl has extremely high initial contact activity, residual activity decreases rapidly and little insecticidal action remains after two days.

Thus, there is a need for a means for producing methomyl in a form where insects will be destroyed without injuring a valuable crop, cotton. In addition, it would be desirable to preserve the high initial contact activity and increase the residual activity of the methomyl. Enhanced residual activity would be valuable for use on any crop, not only on cotton.

According to the instant invention, it has unexpectedly been found that particles of methomyl embedded in a polymer or mixtures of polymers can fulfill all of the above requirements. The composition of the invention is a free-flowing composition comprising 1-50% by weight of methomyl impregnated in 10-98% by weight of a water-insoluble polymer which is soluble in an organic solvent, has an inherent viscosity of at least 0.2 and is capable of releasing methomyl at a rate such that said composition exhibits an undiminished initial activity, an enhanced residual activity and reduced phytotoxic damage to cotton plants relative to solid methomyl.

It should be emphasized that every polymer may not be used for this purpose; only certain polymers permit the passage of sufficient methomyl to have substantially undiminished initial insecticidal activity and enhanced residual activity while exhibiting reduced phytotoxic damage to a sensitive crop such as cotton. Typical of the polymers which may be utilized are poly(methylmethacrylate), poly(ethylmethacrylate), polystyrene and ethyl cellulose.

The polymer must be water-insoluble, have low permeability to water and be capable of dissolving in an organic solvent. The polymer utilized should also have an inherent viscosity of at least about .2 measured for 0.25 g polymer in 50 ml solvent at 20°C

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using a No. 50 Cannon & Fenske viscometer. The particles must also flow freely so they may be readily applied.

DETAILED DESCRIPTION OF THE INVENTION

5 The polymers which may be utilized for the instant invention should have an inherent viscosity of at least about 0.2 and on up to about 2.0. Preferably, at least about 0.4.

10 The polymers which are effective in the instant invention include the following: poly(methylmethacrylate), poly(ethylmethacrylate), methyl methacrylate copolymers with polar monomers, ethyl cellulose, cellulose acetate, cellulose acetatebutyrate, polystyrene, styrene copolymers, poly(vinyl chloride), vinyl chloride-
15 vinyl acetate copolymers and poly(vinyl acetate). There is no intent to limit the polymers to those named above.

 The invention also includes particles consisting of more than one of the above polymers.

20 Inert particulate additives or diluents may be used in conjunction with particles in order to facilitate processing or adjusting the availability of methomyl from the controlled release matrix.

25 Preferred solid diluents include finely divided silicas, kaolinites, diatomites, montmorillonites, hydrous aluminosilicates, calcium carbonates, talcs, crushed brick, pyrophyllites, silicates, etc.

30 The diluent may be present in an amount up to about 50% by weight of the entire composition, preferably up to about 10%. The diluent should be present in an amount of at least about .1%, preferably at least about 2-3%.

 Concerning the particles themselves, the polymer content should be about 10-98 weight %, prefer-

ably 30-95 weight %, and most preferably about 40-90 weight % of the particles. The methomyl content should be in the range of 1-50%, preferably 10-40%, and most preferably 15-35%.

5 The particles may be produced by any of the known methods for making particles with an active ingredient embedded therein, such as co-melting of a polymer and the insecticide followed by grinding the product to a desired particulate size. One method that
10 is advantageous for methomyl is to dissolve the polymer and methomyl in a solvent such as methylene chloride. The solvent is then removed at atmospheric or reduced pressure in an air stream, preferably with some heating. This method permits use of some non-melting polymers or
15 polymers that melt above about 100°C, the decomposition temperature of methomyl. In some cases, an intermediate gel phase may result which can readily be fragmented with slight pressure, reducing the amount of grinding required to obtain a product which is a sufficiently
20 fine powder to be useful in the instant invention. Also, grinding can be facilitated by the use of low molecular weight polymers, or the use of additives that embrittle the polymer such as silica.

 The polymer is then ground by conventional
25 means such as an air mill so that substantially all of the particles are under 300 microns in their longest dimension (at least 90%).

 Generally, it is much preferred that the ground sample be washed with water to remove any unem-
30 bedded methomyl. This should minimize phytotoxic effects of free methomyl on cotton plants. If extremely high initial activity is desired it may be necessary to avoid the wash step in order to leave small amounts of unembedded methomyl in the particulate composition.

Another procedure involves spray-drying the above-mentioned methomyl-polymer solution. Such a procedure is advantageous where grinding is undesirable or uneconomical such as with some relatively soft or low melting polymers.

The preferred method for producing the particle is found in our United States application Serial No. 892,396, filed March 31, 1978, the disclosure of which is incorporated herein by reference.

10 Briefly summarized, the invention in the latter application is as follows:

Initially, methomyl and a polymer are dissolved in one or more organic liquids.

15 The polymer must be water-insoluble, have low permeability to water and be capable of dissolving in an organic solvent, which solvent must also be utilized to dissolve the methomyl. The polymer should also have an inherent viscosity of at least about 0.2. The most preferred polymer is poly(methylmethacrylate). The preferred solvents are methylene chloride, trichloroethylene, ethylene dichloride, and perchloroethylene.

20 The solution is then co-mingled by any conventional high-shear method such as stirring in a Waring blender, colloid mill, or the like with a non-solvent (preferably hexane) or mixture of non-solvents for the methomyl and polymer. Another procedure more suitable for continuous operation is to carry out the co-mingling continuously in a static high turbulence mixer such as a tee or bullet mixer. The solvent must be miscible with the non-solvent. Almost immediately, particles of polymer embedded with methomyl precipitate from the solution. The high-shear agitation is continued during the co-mingling of the non-solvent with the polymer-methomyl solution. The resulting precipitate is then washed to remove solvent and unembedded or free metho-

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myl. The washing step is optional if one desires to have free methomyl for any reason. The particles are recovered by filtration or any other conventional means and dried.

5 The particles are irregular in shape and tend to be under about 300 microns in size in their longest dimension. Typically, at least about 90% of the particles are under 300 microns in their longest dimension and above 5 microns in their shortest.

10 The resulting particles may be applied with particular success to cotton plants wherein phytotoxicity is minimized. Also the initial activity of the controlled release methomyl is quite high. Residual activity of the methomyl-polymer compositions is superior to methomyl alone thereby reducing the number of applications needed to control insect infestation in e.g. cotton plants.

 The following examples are illustrative of the instant invention. Unless otherwise indicated, 20 all parts are by weight and all temperatures in °C.

EXAMPLE 1

 A stirred solution of 16.4 g of methomyl and 40.0 g (Du Pont) Elvacite® 2010 poly(methylmethacrylate) in 100 ml of methylene chloride was treated dropwise 25 with 92 ml of hexane. The stirred solution was added dropwise over a 10 minute period to a solution of 10 g of Emcol® 14 (Witco Chemical Company) surfactant in 500 ml of hexane at -20° and agitated at high speed (> 1,000 rpm) in a high shear Waring blender. The 30 solid particles formed were allowed to settle, the liquid phase was decanted, and the resultant solid was agitated under high shear with 300 ml hexane at 0°, which removes the methylene chloride and hardens the particles. The particulate solid, all of which passed 35 through a 60 mesh USS screen (approx. 250 μ mesh open-

ing) was successively filtered off, dried on a suction funnel, washed with 1 liter of water containing a trace of sodium lauryl sulfate detergent, and dried in an air stream. The product contained about 20% methomyl determined by infrared analysis.

EXAMPLE 2

The process of Example 1 was carried out using high molecular weight Elvacite® 2041 poly(methylmethacrylate). The product contained 21% methomyl and was not quite as fine as that of Example 1.

EXAMPLE 3

The process of Example 1 was carried out using a polymer solution in which was dispersed 2.0 g of Cab-O-Sil® silica (Cabot Corporation), 8.0 g of Elvacite® 2010, 4.1 g methomyl and 100 ml of methylene chloride. The particulate product contained approximately 20% methomyl.

EXAMPLE 4

The process of Example 1 was carried out using polystyrene (Monsanto 314 - Natural) as the polymer. The product contained 18% methomyl.

EXAMPLE 5

The process of Example 1 was carried out using poly(ethylmethacrylate) (Elvacite® 2042) as the polymer. The product contained 17% methomyl.

EXAMPLE 6

The process of Example 1 was carried out using a solution of 10.0 g of poly(vinyl acetate) (Polysciences Company) and 3.75 g of methomyl in 100 ml of methylene chloride in which was stirred 5.0 g Cab-O-Sil® synthetic silica. The product contained 11% methomyl.

EXAMPLE 7

The process of Example 1 was carried out using 5.0 g of poly(methylmethacrylate) (Elvacite® 2010),

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5.0 g polystyrene (Monsanto 314-Natural) and 4.1 g of methomyl in a solution of 100 ml of methylene chloride and 50 ml of hexane. Emcol® 14 (5 g) was used in the cold hexane phase. The product contained 22% methomyl.

5

EXAMPLE 8

The process of Example 1 was carried out using 10.0 g Elvacite® 2010 and 15.0 g methomyl in a solution of 100 ml of methylene chloride and 50 ml of hexane. The product which contained about 30% methomyl was successively extracted with two 250 ml portions of water initially, again after 10 weeks and again similarly extracted after a total of 22 weeks of aging under ambient conditions. The final washing removed less than 0.01 g methomyl. Forced air drying was used after each washing. The final methomyl concentration was 20%.

15

EXAMPLE 9

The process of Example 1 was carried out using ethyl cellulose as polymer (Ethocel®, Dow Chemical Company) in 125 ml methylene chloride. The product contained 17% methomyl.

20

EXAMPLE 10

A solution of 1.0 g poly(methylmethacrylate) (Elvacite® 2010) and 1.5 g methomyl in 20 ml methylene chloride was stripped in a rotary evaporator at 30°.

The solid product was ground to fine powder, sieved through a 50 mesh screen and then washed with about 250 ml of water and dried. Infrared analysis indicated the presence of about 30% methomyl embedded in the polymer.

25

EXAMPLE 11

The process of Example 10 was repeated using polystyrene (Monsanto 314-Natural) as the polymeric material. Approximately 29% methomyl was found in the product.

30

UTILITY

The particulate products of this invention are useful as insecticides and are applied as a dust or water dispersible powder. They are particularly
5 useful for control of insects in cotton.

The cotton plant has many insect pests, each of which contributes to decreasing the yield of seed cotton that can be harvested. Some insects like aphids and plant bugs feed on the sap and retard plant growth.
10 Others like the cotton leaf perforator, beet armyworm and looper, feed on the foliage and reduce the number of bolls that the plant can produce or mature. Still others, like budworms and bollworms, feed on the fruiting body late in the growing season and directly reduce
15 or destroy the harvest. These latter pests are considered by many to be among the greatest insect pests of agriculture today.

Methomyl provides excellent control of a great many insect pests and is particularly effective on the
20 budworm/bollworm complex. When applied directly to cotton, however, some of it is taken up by the cotton leaves and degraded. In a few days, recommended application rates have lost a portion of their effectiveness. Under certain growing conditions, the application of
25 methomyl to most cotton varieties shows a reddening effect from even moderate application rates. Reddening is more likely to occur at high application rates and intensive spray schedules. Growers generally prefer their cotton to exhibit a dark-green appearance.

30 The particles of this invention greatly retard absorption of methomyl by plant foliage, yet provide a ready source to the insect cuticle or stomach. Hence, cotton plants are protected from insect damage with only a minimal opportunity to absorb methomyl. Reduced plant
35 uptake leads to reduced degradation of methomyl and to

protection from reddening of the cotton plants. The methomyl remains available on the plant for insect control for a significantly longer period of time. Use of the particles of this invention leads to improved insect control, reduced cotton reddening and improved residual insecticidal action. Less methomyl is required to achieve a given level of insect control with consequent economy and less dispersal of the insecticide into the biosphere.

10 The particles of this invention readily control pestiferous insects belonging to such orders as Lepidoptera, Homoptera, Hemiptera, Diptera and Coleoptera. More specifically, insects controlled by compositions of this invention include but are not limited to: cotton bollworm (Heliothis zea), tobacco budworm (Heliothis virescens), southern armyworm (Spodoptera eridania), soybean looper (Pseudoplusia includens), beet armyworm (Spodoptera exigua), cotton aphid (Aphis gossypii), tarnished plant bug (Lygus lineolaris),
15 and white flies (Trialeurodes spp. and Bemisia tabaci).
20

 The insects are controlled by applying the particles in any convenient formulation to the locus of infestation, to the area to be protected, or to the pests themselves. For control of insects in agricultural crops, the particle is generally applied to the foliage or other plant parts that are infested or which are to be protected. Effective amounts to be applied depend on the species to be controlled, its life stage, its size and location, the amount of rainfall, the time
25 of year, moisture, temperature, type of application, and other variables. In general, .0625 to 4 kg/ha of the active ingredient may be required for insect control in agriculture with rates of .125 to 2 kg/ha usually
30

being sufficient. Preferred rates for controlling pests in cotton are in the range of .125 to 1 kg/ha.

The particles of this invention may be used as is or may be formulated in conventional ways as dusts, wettable powders, or the like. They may be admixed with diluents and/or surfactants. Suitable diluents are mineral carriers such as clays, talcs, pyrophyllites, hydrous aluminosilicates, fine silicas, etc., and organic carriers like finely divided wood or shell flours. Surfactants such as wetting agents, dispersing agents, antifoam agents and the like may be used alone or in combination, especially if application from an aqueous spray is intended.

Application of the pesticides to plants may be made dry or by spraying from an oil or water carrier. It is often desirable to use surfactants in the oil or water carrier to improve dispersion and wetting, and such surfactants can be added into the formulation or tank-mixed into the spray. Concentrated aqueous dispersions, containing up to 20% of the powders in the spray, or dilute dispersions containing as little as 80 ppm of powder, may be used.

The particles of this invention can be mixed with fungicides, bactericides, acaricides, nematocides, insecticides, or other biologically active compounds in order to achieve desired results with a minimum expenditure of time, effort and material. Amounts of these biologically active materials added for each part by weight of the compound of this invention may vary from about .05 to 25 parts by weight. Suitable agents of this type are well known to those skilled in the art. Some are listed below:

Fungicides:

methyl 2-benzimidazolecarbamate

tetramethyl thiuram disulfide (thiuram)

n-dodecylguanidine acetate (dodine)
manganese ethylenebisdithiocarbamate (maneb)
1,4-dichloro-2,5-dimethoxybenzene (chloroneb)
methyl 1-(butylcarbamoyl)-2-benzimidazolecarbamate

5 (benomyl)

N-trichloromethylthiotetrahydrophthalimide (captan)

N-trichloromethylthiophthalimide (folpet)

Bactericides:

tribasic copper sulfate

10 streptomycin sulfate

Acaricides:

3,3-dimethyl acrylic acid, ester with 2-sec-butyl-4,6-dinitrophenol ("Morocide")

6-methyl-1,3-dithiolo[2,3- β]quinolin-2-one ("Morestan")

15 ethyl 4,4'-dichlorobenzilate (Chlorobenzilate®)

1,1-bis(p-chlorophenyl)-2,2,2-trichloroethane (Kelthane®)

bis(pentachloro-2,4-cyclopentadien-1-yl) (Pentac®)

tricyclohexyltin hydroxide (Plictran®)

Nematicides:

20 S-methyl 1-(dimethylcarbamoyl)-N-(methylcarbamoyloxy)-thioformimidate (Vydate®)

S-methyl 1-carbamoyl-N-(methylcarbamoyloxy)thioformimidate

25 N-isopropylphosphoramidic acid, O-ethyl-O'-[4-(methylthio)-m-tolyl]diester ("Nemacur")

Insecticides:

3-hydroxy-N-methylcrotonamide(dimethylphosphate)ester (Azodrin®)

30 methylcarbamic acid, ester with 2,3-dihydro-2,2-dimethyl-7-benzofuranol (Furadan®)

O-[2,4,5-trichloro- α -(chloromethyl)benzyl]phosphoric acid, O',O'-dimethyl ester (Gardona®)

2-mercaptosuccinic acid, diethyl ester, S-ester with thionophosphoric acid, dimethyl ester (Malathion®)

- phosphorothioic acid, O,O-dimethyl, O-p-nitrophenyl
ester (methyl parathion)
- methylcarbamic acid, ester with α -naphthol (Sevin®)
- methyl O-(methylcarbamoyl)thioacetohydroxamate (methomyl)
- 5 N'-(4-chloro-o-tolyl)-N,N-dimethylformamidine (Galecron®)
- O,O-diethyl-O-(2-isopropyl-4-methyl-6-pyrimidylphosphorothioate (Diazinon®)
- octachlorocamphene (toxaphene)
- O-ethyl O-p-nitrophenyl phenylphosphonothioate (EPN)
- 10 cyano(3-phenoxyphenyl)-methyl-4-chloro- α -(1-methylethyl)-
benzeneacetate (Pydrin®)
- (3-phenoxyphenyl)methyl(\pm)-cis,trans-3-(2,2-dichloro-
ethenyl)-2,2-dimethylcyclopropanecarboxylate
(Ambush®)
- 15 O-ethyl-S-(p-chlorophenyl)ethylphosphonodithioate
(Curacron®)
- phosphorothiolothionic acid, O-ethyl-O-[4-(methylthio)-
phenyl]-S-n-propyl ester (Bolstar®)

EXPERIMENT I

- 20 The foliage of Red Kidney bean plants in the
two-leaf stage (8 days from planting) is sprayed to
runoff with dispersions of the preparations listed be-
low. Dispersions are prepared by stirring appropriate-
ly weighed quantities of the powders in water containing
- 25 sodium lauryl sulfate at 1:5000 and further diluting to
100 ml. After drying, leaves are excised and placed in
covered 10-cm Petri dishes along with moist filter paper
to keep them turgid. Ten southern armyworm larvae were
placed in each dish. The test units are kept in a room
- 30 maintained at $77^{\circ}\pm 2^{\circ}\text{F}$ and $55\pm 5\%$ R.H. Results are recor-
ded at the end of two days and are listed below.

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	<u>Treatment</u>	<u>Spray Concentration (ppm) Active Ingredient</u>	<u>% Mortality (2 days)</u>
	Methomyl Control	100	5
5	Product of Example 1 (20% Active)	100	100
	Product of Example 10 (30% Active)	100	100
	Product of Example 11	100	100
10	Untreated	-	-

EXPERIMENT II

An experiment is performed that is similar in all respects to Experiment I except that the leaves are excised from the plants and fed to southern armyworm larvae at various intervals rather than immediately. Results are evaluated two days later.

Treatment	Active Ingredient	Spray Concentration (ppm)				% Mortality (days)			
						2	5	7	9
20 Methomyl Control	100					0-15	0	0	0
Product of:									
Example 1	100					100	-	93	90
	50					-	90	-	-
Example 2	100					100	95	-	-
25 Example 3	100					90	-	-	-
Example 4	100					90	-	-	-
Example 5	100					95	-	90	-
Example 6	100					70	-	-	-
Example 7	100					100	-	-	-
30 Example 8	100					100	-	100	-
	50					100	-	100	-
Example 9	100					80	-	-	-
Example 10	100					100			
Example 11	100					100			
Untreated	-					0			

EXPERIMENT III

Fifty ml portions of dispersions of the test material prepared by the procedure described in Experiment I are poured over 10 southern armyworm larvae placed in a Buchner funnel (8 cm in diameter). The larvae are then transferred using forceps to Petri dishes containing moistened filter paper and an excised bean leaf for food. Results are read two days later.

<u>Treatment</u>	<u>Concentration (ppm)</u>	<u>% Mortality (2 days)</u>
10 Product of Example 1	100	100
Untreated	-	-

EXPERIMENT IV

An experiment is performed that is similar in all respects to that described in Experiment I except that the excised leaves are fed to soybean loopers. Excellent control is obtained.

<u>Treatment</u>	<u>Spray Concentration (ppm)</u> <u>(Active Ingredient)</u>	<u>% Mortality (2 days)</u>
Product of	200	100
20 Example 1	100	80

EXPERIMENT V

Potted cotton plants approximately 25 cm high having 3-4 true leaves are sprayed to run-off with aqueous dispersions of compositions of this invention at 500 ppm. The sprays contain sodium lauryl sulfate at a concentration of 1:5000. Another set of plants is similarly treated with methomyl. After drying, plants are set out in the greenhouse and held for observation.

<u>Treatment (500 ppm AI)</u> ^{1/}	<u>Rating</u> ^{2/}	
	<u>7 days</u>	<u>8 days</u>
Product of:		
Example 1	trace R	-
Example 2	trace R	-

	Example 4	0.5R	-
	Example 5	-	0.2R
	Example 6	-	0.2R
	Example 10	-	0.2R
5	Methomyl Control	3R	-
	Untreated	0	-

1/ AI = Active ingredient.

2/ R denotes typical methomyl effect, i.e., reddening of older leaves, slight puckering and black stippling of younger leaves.

10 Rating is on the basis of 0-10, with 10 indicating total leaf area involvement.

Claims:

1. An insecticidal particulate composition comprising methomyl and a solid carrier characterised in that said composition is a free-flowing composition comprising 1-50% by weight of methomyl impregnated in 10-98% by weight of a water-insoluble polymer which is soluble in an organic solvent, has an inherent viscosity of at least 0.2 and is capable of releasing methomyl at a rate such that said composition exhibits an undiminished initial activity, an enhanced residual activity and reduced phytotoxic damage to cotton plants relative to solid methomyl.
2. The composition of claim 1 in which the polymer is poly(methylmethacrylate).
3. The composition of claim 1 in which the polymer is polystyrene.
4. The composition of claim 1 in which the polymer is ethyl cellulose.
5. The composition of claim 1 in which the polymer is poly(ethylmethacrylate).
6. The composition of claim 1 in which the polymer is poly(vinyl acetate).
7. A composition of any of claims 1-6 in which said polymer is diluted with 1-50% by weight of a diluent, based on the whole composition.
8. The composition of any of claims 1-7 in which the methomyl content is 10 to 40% by weight and the polymer content is 30-95% by weight.
9. The composition of any of claims 1-8 in which at least 90% of the particles are under 300 microns in their longest dimension.
10. The composition of any of claims 1-9 which has been washed to remove methomyl from the surface of the polymer particles.
11. The particulate composition of any of claims 1-10 in admixture with a surfactant and/or a solid or

liquid diluent.

12. A method of making the composition of claim 1 which comprises comingling a solution of methomyl and said polymer in an organic solvent with a non-solvent for methomyl and said polymer, under conditions of high shear, and recovering free-flowing particles comprising methomyl and said polymer.

13. The method of claim 12 wherein said solvent is methylene chloride, trichloroethylene, ethylene chloride or perchloroethylene and said non-solvent is hexane.

14. The method of claim 12 or 13 wherein the particulate product is washed to remove methomyl from the surface of the polymer particles.

15. A method for controlling an insect pest by applying to the locus of said pest an effective amount of an insecticidal composition characterised in

that said insecticidal composition is the composition of any of claims 1-11.

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The composition may be made by precipitating the components from solution by comingling with a non-solvent under conditions of high shear.

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EUROPEAN SEARCH REPORT

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DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. ²)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
D	<u>CA - A - 786 777 (DU PONT)</u> * Claims 1,7,8; page 3, lines 12-16,29,30; page 12, lines 5-9; page 21, example 1; pages 23,24; example 15 * --	1,7-9 11,15	A 01 N 9/12 C 07 C 149/00
	<u>CH - A - 483 196 (SHELL)</u> * Claim 6; column 8, lines 29-35 * --	1,3	TECHNICAL FIELDS SEARCHED (Int.Cl. ²)
	<u>DE - A - 1 936 748 (CIBA)</u> * Claims 1,2; page 22, line 6; page 7, lines 2-5 * --	1-6	A 01 N 9/12 17/08
	<u>CH - A - 430 660 (THE NATIONAL CASH REGISTER COMP.)</u> * Column 1, lines 4-24; column 2, lines 20-34; page 4, lines 1,2 * ----	12,13	CATEGORY OF CITED DOCUMENTS X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons &: member of the same patent family, corresponding document
<div style="display: flex; align-items: center;"> <div style="border: 1px solid black; padding: 2px; margin-right: 10px;"> </div> <div> The present search report has been drawn up for all claims </div> </div>			
Place of search	Date of completion of the search	Examiner	
The Hague	11-07-1979	DECORTE	